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Generation of femtosecond laser pulses at 263 nm by $K_3B_6O_{10}Cl$ crystal*

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The third harmonic generation (THG) of a linear cavity Ti:sapphire regenerative amplifier by use of a $K_3B_6O_{10}Cl$ (KBOC) crystal is studied for the first time. Output power up to 5.9 mW is obtained at a central wavelength of 263 nm, corresponding to a conversion efficiency of 4.5% to the second harmonic power. Our results show a tremendous potential for nonlinear frequency conversion into the deep ultraviolet range with the new crystal and the output laser power can be further improved.

Keywords: nonlinear optical crystal, KBOC, third harmonic generation, femtosecond ultraviolet

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1. Introduction

Ultraviolet (UV) and deep ultraviolet (DUV) femtosecond laser pulses have been extensively employed in the research of ultrafast photophysical processes in atoms, molecules and clusters.[1–3] Such sources are also key ingredients for time-resolved photoelectron spectroscopy, especially $\sim$ 180 nm for the time-resolved angle resolved photoemission spectroscopy (ARPES).[4,5] However, based on the frequently used borate family nonlinear optical (NLO) crystals like LiB_3O_5 (LBO),[6] β-BaB_2O_4 (BBO),[7] $CsLiB_6O_{10}$ (CLBO),[8] and BiB_3O_6 (BIBO),[9] the second harmonic generation (SHG) usually achieves the UV in the spectral range of 300 nm–400 nm. The $KBe_2BO_3F_2$ (KBBF)[10] has the power to generate DUV pulses shorter than 180 nm through SHG directly,[11,12] while its layered structure imposes restrictions on the wide application. To further shorten the wavelength, a more sophisticated frequency up-conversion technique is required, such as sum frequency generation (SFG). For example, the generation of femtosecond pulses in a wavelength range between 172.7 nm and 187 nm based on SFG in LBO was presented by Seifert *et al.* in 1994.[13] Nevertheless, a temperature control system is commonly needed due to the defect that the birefringence of LBO is sensitive to the temperature. In 2015, Kumar *et al.* obtained an ultraviolet source at 355 nm in BIBO based on SFG of a mode-locked Yb-fiber laser at 1064 nm.[14] The research on new NLO crystals with excellent NLO response has been enthusiastically carried out for DUV pulses generation by employing the SFG technique. Recently, using $Ba_{1-x}B_{2-y-z}O_4Si_xAl_yGa_z$ ($x = 0 \sim 0.15$, $y = 0 \sim 0.01$, $z = 0 \sim 0.04$, $x^2+y^2+z^2 \neq 0$, BBSAG) crystal, DUV pulses with high repetition in a tunable range of 192.5 nm–210 nm was demonstrated by Meng *et al.*[15] In 2015, a novel high quality NLO crystal called $K_3B_6O_{10}Cl$ (KBOC) with size up to 35 mm×35 mm ×11 mm was presented by Wu *et al.*[16] Prior to this report, a series of studies on the properties of KBOC has been performed, such as the structure and NLO properties,[17] the influence of pressure on the SHG tensor[18] and the electronic, elastic, piezoelectric, acoustic, and Raman spectroscopic properties.[19,20] However, the SFG from KBOC has not been reported.

As a new negative uniaxial crystal, KBOC crystal possesses unique NLO properties for frequency up-conversion in the visible and UV. Its spectral transmittance extends from 3460 nm in the infrared down to 180 nm in the UV region[21] as shown in Table 1, which is 10-nm shorter than that of BBO crystal in the UV cut-off. In particular, the KBOC combines the non-hygroscopic property differentiated from BBO, CLBO, and LBO with high optical damage threshold, which suggests that it could ensure steady long-term operation for large national laser facilities.[22] Besides, the temporal walk-off lengths of the crystals between the fundamental frequency (FF) and the third harmonic (TH) are calculated according to the pulse duration of 35 fs in Table 1. It can be obtained

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that the walk-off length of KBOC is much longer than those of BBO, CLBO, and LBO. The intensity of the TH is proportional to the square of the product of the effective nonlinear coefficient and the length of the crystal. Accordingly, it may compensate for the small effective nonlinear coefficient in the nonlinear frequency conversion process and has access to achieving a considerable conversion efficiency. The combination of these excellent properties makes KBOC an attractive nonlinear crystal for frequency up-conversion in the DUV spectral region.

Table 1. NLO characteristics of KBOC, BBO, CLBO, and LBO for type-I THG at 800-nm wavelength.

Crystal	UV cut-off/nm	Phase-matching angle/(°)	Temporal walk-off length/(mm)@35 fs	d_{eff} /(pm/V)	Hygroscopicity	Damage threshold/(GW/cm^2)
KBOC	180	75.4	0.84	0.22	none	9@5 ns, 1 Hz, 1064 nm
BBO	190	44.3[23]	0.05	1.87	slight	13.5@1 ns, 10 Hz, 1064 nm
CLBO	180	56.1[23]	0.08	0.75	high	26@1 ns, 10 Hz, 1064 nm
LBO	160	$\theta = 90°$ $\varphi = 77.4°$[23]	0.08	0.21	slight	19@1.3 ns, 1064 nm

Nevertheless, many of the vital nonlinear optical properties of KBOC have not yet been entirely investigated, resulting in the limit of the optimum application of this crystal for nonlinear frequency up-conversion. We have studied the second harmonic (SH) properties of a femtosecond Ti:sapphire amplifier with KBOC crystal generating 220-mW pulses at 396 nm.[22] In this paper, we investigate experimentally the third harmonic generation (THG) of a cavity mode size adjustable Ti: sapphire femtosecond regenerative amplifier[24] for the first time in KBOC. The highest THG conversion efficiency of 4.5% corresponding to the SH power is obtained with a 1-mm-long KBOC crystal and the maximum output power is 5.9 mW at the central wavelength of 263 nm. The results provide a reference for the SFG characteristic of KBOC. Coupled with the remarkable properties and the large size, the crystal will have novel applications in various areas.

2. Experimental setup

As shown in Fig. 1, using the Sellmeier equations of KBOC derived by Wu *et al.*,[16] we calculated the type-I phase-matching angles for SHG and THG and the corresponding walk-off angles in the THG process versus fundamental wavelength. We can obtain from Fig. 1 that the shortest fundamental wavelength for THG is about 777 nm and the walk-off angles between the TH (extraordinary wave) and the fundamental frequency (FF, ordinary wave) is $< 1°$ at 800 nm. The variation tendency of the walk-off angles of THG is similar to that of SHG described in Ref. [22].

A schematic of the experimental setup is shown in Fig. 2. The pump laser was a cavity mode size adjustable femtosecond Ti:sapphire regenerative amplifier at 1 kHz which was described in detail elsewhere.[24] It is able to produce 4.8-mJ average power near 800 nm with the pulse duration of 35 fs. The spot diameter of the FF light through the 5:1 Galilean telescope was reduced to 5 mm in a 0.5-mm-long KBOC crystal cut at $\theta = 42.7°$ and $\varphi = 30°$ for type-I phase-matching. By employing a dichroic mirror (DM) which was high reflection-coated in a range between 370 nm and 430 nm and anti-reflection-coated between 750 nm and 850 nm, the SH light was separated from the residual FF light. A half-wave plate (HWP) was used to rotate the polarization of the FF light before the THG stage. The spot diameters of the two beams were both halved, passing through the 2:1 Galilean telescope. The delay between FF and SH was controlled by the delay line (DL). The THG was achieved by collinear SFG between the FF and the SH in a 1-mm-long KBOC cut at $\theta = 75.4°$ and $\varphi = 30°$ for type-I phase-matching. The photograph of the two KBOC crystals is shown in the inset of Fig. 2. According to the equation in Ref. [22], the effective nonlinear coefficient of the second crystal was calculated to be about 0.22 pm/V. It should be noted that neither of the two crystals used in this work is coated.

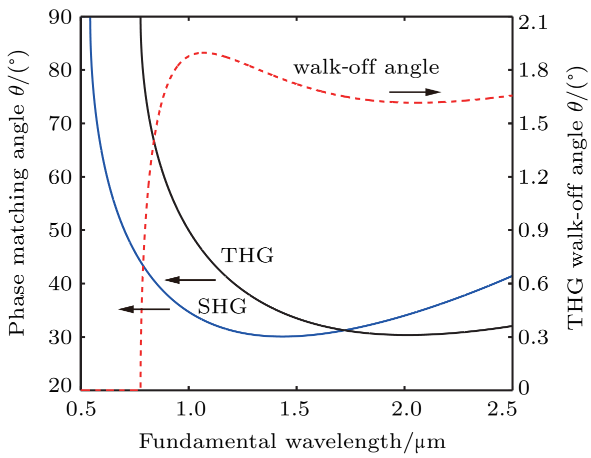

Fig. 1. (color online) Curves of walk-off angle for THG and type-I phase-matching angles for SHG and THG versus fundamental wavelength.

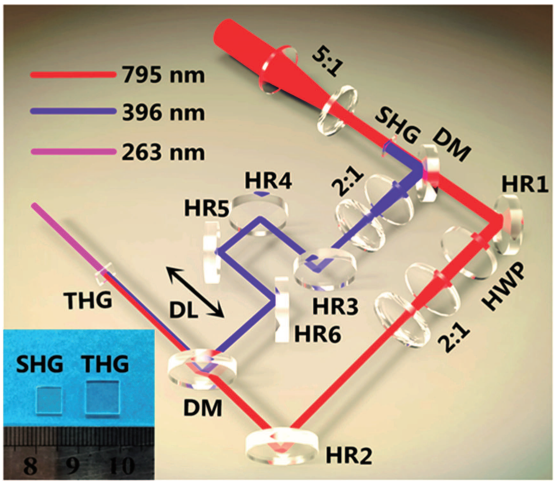

Fig. 2. (color online) Schematic diagram of the experimental setup. 5:1 and 2:1: Galilean telescopes, DM: dichroic mirror, HR1 and HR2: 45° high reflection-coated mirrors for FF light, HR3-HR6: 45° high reflection-coated mirrors for SH light, HWP: half-wave plate, DL: delay line for FF-SH temporal overlap. Inset shows the crystals used in the experiment.

3. Results and discussion

The plots of output power of SH at 396 nm and the corresponding conversion efficiency of SHG versus the input power of FF are shown in Fig. 3(a). The maximum output power of 131 mW is obtained at the FF power of 500 mW and the highest SHG conversion efficiency is about 31% for 350 mW of input FF power. With the increase of input power, the conversion efficiency tends to be saturated and slightly decline which could account for the improper length of the crystal. The tested transmissivities of a 1-mm-long uncoated KBOC at 800 nm and 396 nm are both less than 90%. While the optimum thickness of the KBOC crystal for the 35-fs FF pulse is calculated to be 0.3 mm according to the temporal walk-off theory. By using an anti-reflection-coating to minimize the loss of reflection and a thinner crystal to weaken the group velocity mismatch effect, the SH conversion efficiency may be further improved.

The output power and the conversion efficiency for the THG each as a function of SH power are shown in Fig. 3(b). The output power of TH reaches 5.9 mW with SH input power of 131 mW and the highest THG conversion efficiency of 4.5% is obtained. As mentioned above, the transmissivity of the uncoated KBOC is one of the most important factors that influence the TH output power and conversion efficiency. Another key reason for this may account for the limitation of the phase-matching wavelength. As can be seen clearly from Fig. 1, the shortest wavelength of FF for THG is about 777 nm, therefore a part of the FF power cannot be utilized effectively in the SFG process. In addition, the internal phase-matching angle varies from 90° to 68.3° in a wavelength range of 777 nm–830 nm and the difference in phase-matching angle is as large as 21.7°. Consequently, the dramatic variation of the internal phase-matching angle for THG near 800 nm limits the THG phase-matching bandwidth, resulting in the restrictions on not only TH output power and conversion efficiency but also the TH spectrum bandwidth shown in Fig. 4. However, it should be notable that there is no obvious saturation tendency of the THG conversion efficiency, indicating that it still has the potential to be ameliorated with the increase of the input power of SH.

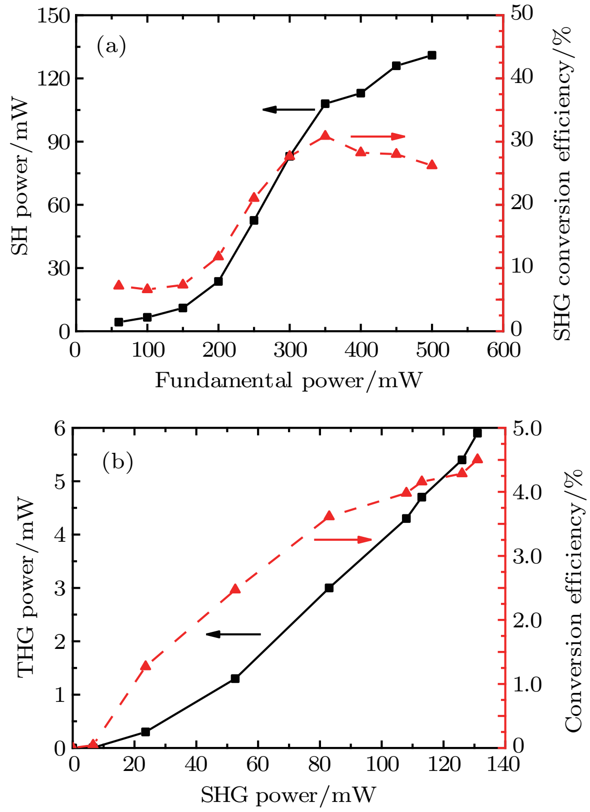

Fig. 3. (color online) (a) Plots of SH power and SHG conversion efficiency versus fundamental power and (b) plots of THG power and conversion efficiency versus SHG power.

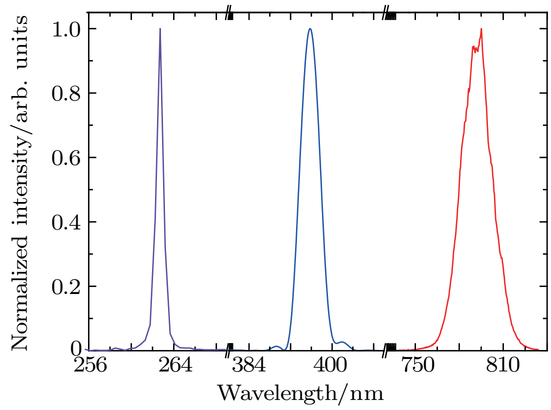

Fig. 4. (color online) Typical spectra of the FF, SH, and TH pulses.

Figure 4 shows the typical spectra of the FF, SH, and TH pulses measured with a spectrometer (Ocean Optics, USB2000+XR1). The central wavelengths are at 795 nm, 396 nm, and 263 nm, respectively. The full width at half maximum (FWHM) bandwidth of the TH spectrum is estimated to be ∼ 0.8 nm, limited by the instrument resolution. The temporal profile of the SH pulse is characterized by a home-made dispersion-free transient-grating frequency-resolved optical gating (TG-FROG) with a 0.05-mm-thick silica plate as the nonlinear medium. The experimental and retrieved TG-FROG traces for the uncompressed SH pulses are shown in Figs. 5(a) and 5(b), while figures 5(c) and 5(d) illustrate the retrieved spectral and temporal intensity and phase profiles, respectively. The FWHM of the spectrum is about 4.4 nm with a central wavelength at 395.7 nm and the corresponding transform limited (TL) pulse width is 39 fs shown with the dash curve in Fig. 5(d). Without chirp compensation, the SH pulse duration is calculated to be about 71 fs.

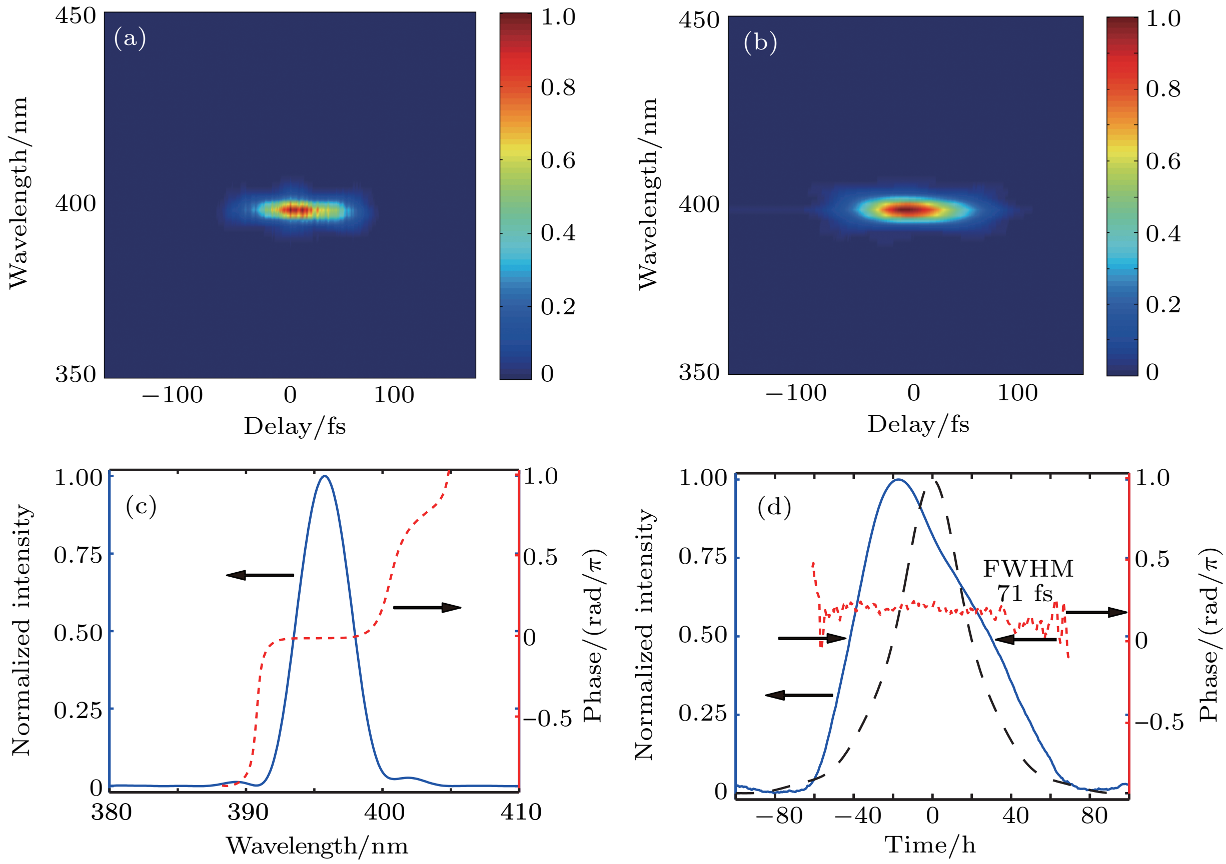

Fig. 5. (color online) (a) Measured and (b) retrieved TG-FROG traces of the SH pulse. Intensity and phase profiles in (c) frequency domain and (d) time domain. The dash curve in panel (d) is the TL pulse width of SH.

4. Conclusions

In this work, we first demonstrate the generation of DUV pulses at 263 nm from a cavity mode size adjustable Ti:sapphire femtosecond regenerative amplifier by using a novel nonlinear KBOC crystal through two stages of SHG and SFG. In the SHG process, UV pulse at 396 nm of 71 fs characterized by a TG-FROG with the maximum output power of 131 mW and the highest conversion efficiency of 31% is obtained in a 0.5-mm-thick KBOC crystal. After the subsequent SFG, DUV pulses with the maximum output power of 5.9 mW and a conversion efficiency of 4.5% versus the SH power were achieved. The experimental results indicate that the DUV pulse output power and conversion efficiency still have the feasibility to be improved with higher SH power and an anti-reflection-coated KBOC crystal for FF, SH, and TH.

References

[1] Mondal S and Puranik M 2016 *Phys. Chem. Chem. Phys.* **18** 13874

[2] Yang C F, Su H M, Sun X D and George M W 2012 *J. Chem. Phys.* **136** 204507

[3] Hertel I V and Radloff W 2006 *Rep. Prog. Phys.* **69** 1897

[4] Mou D X, Sapkota A, Kung H H, Krapivin V, Wu Y, Kreyssig A, Zhou X J, Goldman A I, Blumberg G, Flint R and Kaminski A 2016 *Phys. Rev. Lett.* **116** 196401

[5] Mou D X, Konik R M, Tsvelik A M, Zaliznyak I and Zhou X J 2014 *Phys. Rev. B* **89** 201116

[6] Chen C T, Wu Y C, Jiang A D, Wu B C, You G M, Li R K and Lin S J 1989 *J. Opt. Soc. Am. B* **6** 616

[7] Chen C T, Wu B C, Jiang A D and You G M 1985 *Sci. Sin. B* **28** 235

[8] Mori Y, Kuroda I, Nakajima S, Sasaki T and Nakai S 1995 *Appl. Phys. Lett.* **67** 1818

[9] Hellwig H, Liebertz J and Bohaty L 1999 *Solid State Commun.* **109** 249

[10] Mei L F, Wang Y B, Chen C T and Wu B C 1993 *J. Appl. Phys.* **74** 7014

[11] Togashi T, Kanai T, Sekikawa T, Watanabe S, Chen C T, Zhang C Q, Xu Z Y and Wang J Y 2003 *Opt. Lett.* **28** 254

[12] Dai S B, Zong N, Yang F, Zhang S J, Wang Z M, Zhang F F, Tu W, Shang L Q, Liu L J, Wang X Y, Zhang J Y, Cui D F, Peng Q J, Li R K, Chen C T and Xu Z Y 2015 *Opt. Lett.* **40** 3286

[13] Seifert F, Ringling J, Noack F, Petrov V and Kittelmann O 1994 *Opt. Lett.* **19** 1538

[14] Kumar S C, Bautista E S and Ebrahim-Zadeh M 2015 *Opt. Lett.* **40** 403

[15] Meng X H, Liu H G, Huang J H, Dai S T, Deng J, Ruan K M, Chen J M and Lin W X 2015 *Acta Phys. Sin.* **64** 164205 (in Chinese)

[16] Wu H P, Yu H M, Yang Z H, Han J, Wu K and Pan S L 2015 *J. Materiomics* **1** 221

[17] Wu H P, Pan S L, Poeppelmeier K R, Li H Y, Jia D Z, Chen Z H, Fan X Y, Yang Y, Rondinelli J M and Luo H S 2011 *J. Am. Chem. Soc.* **133** 7786

[18] Wang H, Kong L Y, Zhao X Y, Lv Z L, Li T W, Ju W W, You J H and Bai Y 2013 *Appl. Phys. Lett.* **103** 101902

[19] Han H, Yin G and Wickramaratne D 2013 *Comput. Mater. Sci.* **69** 81

[20] Gong X Y, Zhao X Y, Lv Z L, Li T W, You J H and Wang H 2014 *Comput. Mater. Sci.* **83** 86

[21] Wu H P, Pan S L, Yu H W, Jia D Z, Chang A M, Li H Y, Zhang F F and Huang X 2012 *Cryst. Eng. Commum.* **14** 799

[22] Zhang N H, Teng H, Huang H D, Tian W L, Zhu J F, Wu H P, Pan S L, Fang S B and Wei Z Y 2016 *Chin. Phys. B* **25** 124204

[23] Smith A V, "SNLO", http://www.as-photonics.com/snlo

[24] He P, Teng H, Zhang N H, Liu Y Y, Wang Z H and Wei Z Y 2016 *Acta Phys. Sin.* **65** 244202 (in Chinese)